Norman. F.H. Bright

CANADA

DEPARTMENT OF MINES AND TECHNICAL SURVEYS

MINES BRANCH

OTTAWA

MINERAL SCIENCES DIVISION

INTERNAL REPORT MS-64-57

EXPERIMENTS IN THE Au-Bi-Te SYSTEM

by

Ernst W. Winkler and Norman F.H. Bright

June 29, 1964

Crown Copyright Reserved

Prepared for Publication in "Solid State Communications"

Note: For Publication in "Solid State Communications"

MINES BRANCH

Mineral Sciences Division

REPORT COPY DISPOSAL SHEET

(Author) E. W. Winkler and M.F.H. Bright

TITLE: Experiments in the An-Bi-Te System

(Date) July 3, 1964

Internal Report No. MS-64-57

(9 cc.)

Copy No.	Sent to:	Date	By
1	Dr. B.N. Brockhouse, Editor, "Solid State Communications"		
2	Dept. of Physics, McMaster University, Hamilton, Ont.		
3	Dr. John Convey, Director, Mines Branch		
4	" " " "		
5	Dr. A.T. Prince		
6	Dr. M.F.H. Bright		
7	Dr. E.W. Winkler, P.M.D.		
8	Phys. Chem. Sect. Files		
9	Reports Secretary		
10			
11			
12			
13			
14			
15			
16			
17			
18			
19			
20			
21			
22			
23			
24			
25			

cc: N.F.H. Bright
E.W. Winkler
Director's Files
Physical Chemistry Section files
Min. Sci.Div. Repts. Sect.
MSD files

NFHB-8?

90-04691/

Mineral Sciences Division,
A.T. Prince, Chief

555 Booth Street,
Ottawa 4, Ontario.
July 9, 1964.

Professor B.N. Brockhouse,
Editor, "Solid State Communications",
Department of Physics,
McMaster University,
Hamilton,
Ontario.

Dear Dr. Brockhouse:

Enclosed with this letter are two copies of a manuscript entitled "Experiments in the Au-Bi-Te System" by Ernst W. Winkler and myself, which we wish to submit through you, as the Canadian editor, for publication in "Solid State Communications". You will observe that the "second language long title" is given in German on page 1 of the manuscript, the text of the paper proper commencing on page 2. You will observe also that, since this is a publication emanating from a Government establishment, Crown Copyright is reserved on any subsequent reproduction of this paper. We trust that you will find this contribution to be in the correct format and suitable for publication. We should appreciate being advised of your safe receipt of this paper.

Yours very truly,

Norman F.H. Bright,
Head,
Physical Chemistry Section.

Mineral Sciences Division Report MS-64-57

EXPERIMENTS IN THE Au-Bi-Te SYSTEM

by

Ernst W. Winkler* and Norman F.H. Bright**

- - - -

ABSTRACT

The system Au-Bi-Te has been studied by thermal analysis, X-ray diffraction, DTA and metallographic polished section techniques. The join Au-Bi_2Te_3 has a eutectic point at 67.7 mol.% Au:32.3 mol.% Bi_2Te_3 and 472°C. The join $AuTe_2$ - Bi_2Te_3 has a eutectic point at 78.1 mol.% $AuTe_2$:21.9 mol.% Bi_2Te_3 and 408°C. A ternary eutectic occurs at 12% Au, 86% Bi, 2% Te and 210°C. The system contains no ternary compound. Seebeck coefficient, electrical conductivity, hardness and Hall coefficient measurements are being made.

Das System Au-Bi-Te wurde mittels thermischer Analyse, Röntgenaufnahmen, differentieller Thermoanalyse und Metallschliffe untersucht. Der Schnitt Au-Bi_2Te_3 zeigt bei 67.7 mol.% Au:32.3 mol.% Bi_2Te_3 ein bei 472° schmelzendes Eutektikum. Im Schnitt $AuTe_2$ - Bi_2Te_3 liegt das Eutektikum bei 78.1 mol.% $AuTe_2$:21.9 mol.% Bi_2Te_3 und 408°. Ein ternäres Eutektikum tritt bei 12% Au, 86% Bi und 2% Te und 210°C auf. Es wurde keine ternäre Verbindung gefunden. Seebeck-koeffizienten, elektrische Leitfähigkeit, Härte und Hallkoeffizienten werden bestimmt.

---

* National Research Council of Canada Postdoctorate Research Fellow assigned to the Mines Branch.

** Head, Physical Chemistry Section, Mineral Sciences Division, Mines Branch, Department of Mines and Technical Surveys, Ottawa, Canada.

The system Au-Bi-Te is of technological importance in view of the use of bismuth telluride in components of thermoelectric refrigeration devices and the employment of gold as a connecting link between such components. The system also has mineralogical significance in that both gold and bismuth occur naturally as tellurides. The binary systems Au-Bi, Au-Te and Bi-Te are reasonably well established[1]; the compounds Bi_2Te_3 and $AuTe_2$ are both stable compounds with congruent melting points and well-established physical and chemical properties[2]. There is, however, virtually no information concerning phase relationships in the ternary system, and particularly in that portion of the system involving Bi_2Te_3 as one of the stable phases. A very limited solid solubility of Au in Bi_2Te_3 has been claimed[3].

The phase relations have been studied using thermal analysis to give liquidus and solidus temperatures, DTA to give supplementary information on melting and transition points, X-ray diffraction for identification of phases, and metallographic polished sections to give information concerning the number of phases and physical structure of the assemblages (e.g. eutectic characteristics, etc.). Throughout this study, either the elements of 99.999% purity or pre-formed compounds made from them were used.

All heating of compositions was done in quartz or Vycor tubes in the absence of oxygen in order to prevent oxidation of either the bismuth or the tellurium. Since the partial pressure of tellurium above all compositions in this system is fairly high, precautions were taken to ensure that no loss of tellurium occurred from a composition by arranging that, in the sealed tube in which it was prepared, the portion containing the sample itself was always a few degrees cooler

than the remainder of the tube. It was found possible to prepare, by this technique, exactly stoichiometic Bi_2Te_3 and $AuTe_2$, as shown by chemical analysis, the examination of metallographic polished sections, X-ray diffraction examination, and the absence of uncombined, condensed tellurium.

The join Au-Bi_2Te_3 was found to be a true binary join with a eutectic composition at 67.7 mol.% Au:32.3 mol.% Bi_2Te_3 and a melting point of 472°C. The liquidus and solidus relations in this sub-system are shown in Figure 1. Very limited solid solubility of Au in Bi_2Te_3 may occur at temperatures near the liquidus but exsolution occurs on annealing at lower temperatures. The join $AuTe_2$ - Bi_2Te_3 was also found to be a true binary join with a eutectic composition at 78.1 mol.% $AuTe_2$:21.9 mol.% Bi_2Te_3 and a melting point of 408°C. The compound $AuTe_2$ was found to have a melting point of 469°C with a crystalline transition at 435°C. This sub-system is shown in Figure 2.

The join Au_2Bi - Bi_2Te_3 is suspected also to be true binary, although work on this sub-system is not complete and may be complicated by a possible incongruity of melting of the compound Au_2Bi. A ternary eutectic composition of melting point 210°C and composition 12% Au: 86% Bi: 2% Te occurs in the compatibility triangle Bi_2Te_3 - Bi - Au_2Bi. No evidence of the formation of any ternary Au-Bi-Te compound has been found anywhere in this system.

Work is in progress to complete the phase diagram of the ternary system with the establishment of other ternary eutectic compositions and the liquidus isotherms.

Measurements of the Seebeck coefficient along the join $AuTe_2$-Bi_2Te_3 have been made and will be reported elsewhere later. Similarly, Seebeck

coefficient measurements along the join Au-Bi_2Te_3 are being made but have been complicated by the occurrence of segregation in the crystallization of these compositions. Measurements of the hardness, electrical conductivity and Hall coefficient of selected compositions in the system will be made and reported elsewhere when complete.

REFERENCES

1. HANSEN M., "Constitution of Binary Alloys", pp 188-189, 234 and 339. McGraw-Hill Book Company (1958).

2. BRODY A. and LEVIN B., Intern. Journ. Phys. Chem. Solids, 23, 1597 (1962).

3. KEYS J.D. and DUTTON H.M., Journ. Appl. Phys., 34, 1830 (1963).

FIGURES

Figure 1. The Au - Bi_2Te_3 Join.

Figure 2. The $AuTe_2$ - Bi_2Te_3 Join.

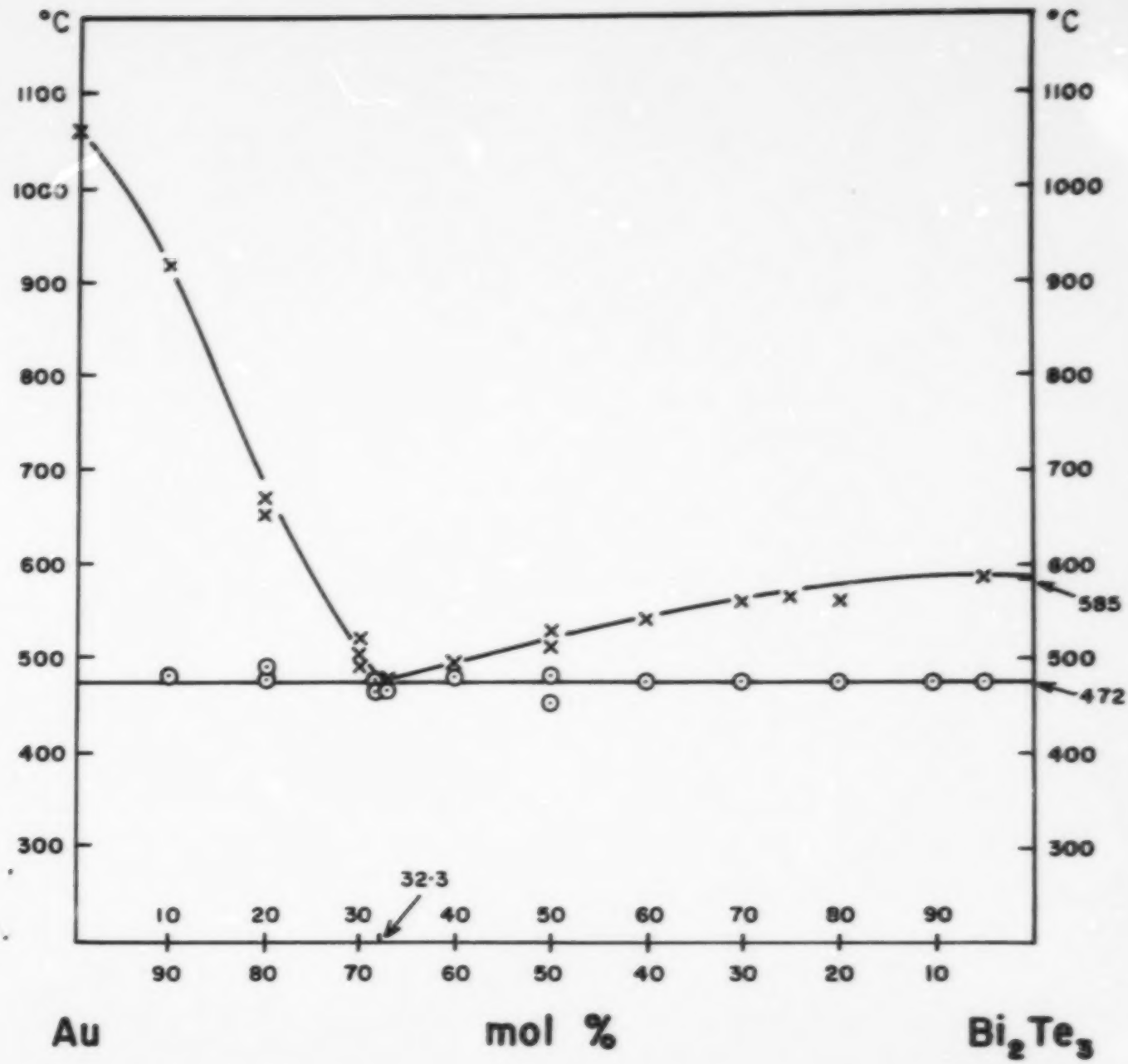
°C
1100
1000
900
800
700
600
500
400
300
585
472
32·3
10
20
30
40
50
60
70
80
90
Au
mol %
Bi2Te3

Figure 1. The Au - $Bi_2 Te_3$ Join.

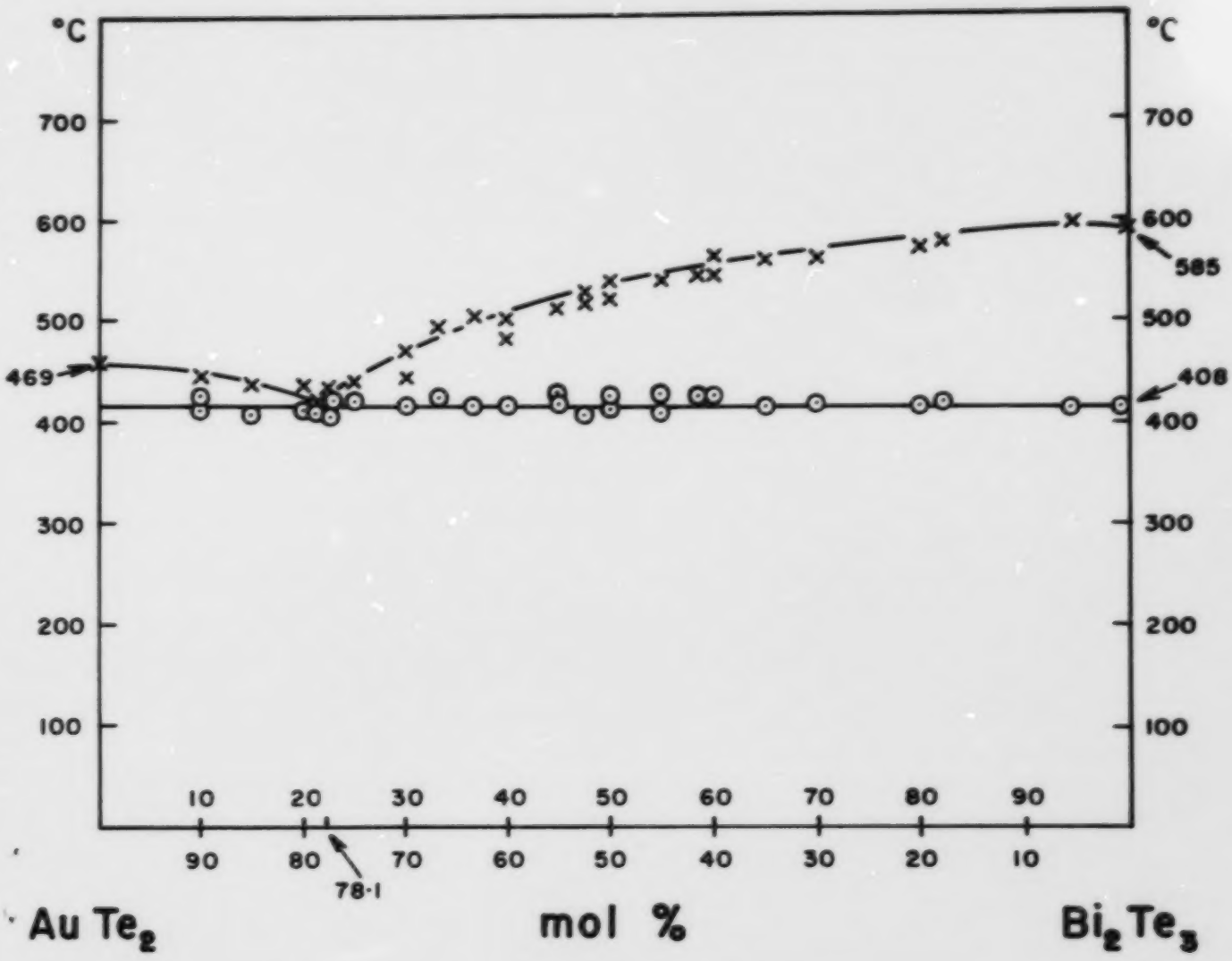
°C
700
600
500
400
300
200
100
469
585
408
10
20
30
40
50
60
70
80
90
78·1
Au Te2
mol %
Bi2 Te3

Figure 2. The $AuTe_2$ - Bi_2Te_3 Join.